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INFRARED HETERODYNE SPECTROSCOPY OF ASTRONOMICAL & LABORATORY SOURCES AT 8.5 μm

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M. MUMMA
T. KOSTIUK
S. COHEN
D. BUHL
P. C. VON THUNA

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"Infrared Heterodyne Spectroscopy of Astronomical
and Laboratory Sources at $8.5\ \mu\text{m}$ "

by

M. Mumma
T. Kostiuks

Astrochemistry Branch
Laboratory for Extraterrestrial Physics

S. Cohen
Laser Technology Branch
NASA/Goddard Space Flight Center
Greenbelt, Maryland 20771

D. Buhl
National Radio Astronomy Observatory
Greenbank, West Virginia

and

P. C. von Thuna
Arthur D. Little, Inc.
Cambridge, Massachusetts

ABSTRACT

The first infrared heterodyne spectrometer using tuneable semiconductor (PbSe) diode lasers has been constructed and was used near $8.5\ \mu\text{m}$ to measure absorption line profiles of N_2O in the laboratory and black body emission from the Moon and from Mars. Spectral information was recorded over a 200 MHz bandwidth using an 8-channel filter bank. The resolution was 25 MHz ($8.3 \times 10^{-4}\ \text{cm}^{-1}$) and the minimum detectable (black body) power was 1×10^{-16} watts for 8 minutes of integration. The results demonstrate the usefulness of heterodyne spectroscopy for the study of remote and local sources in the infrared.

*National Academy of Sciences/Resident Research Associate

The first successful infrared heterodyne spectrometer featuring semi-tuneable semiconductor diode lasers was constructed and used near $8.5\text{ }\mu\text{m}$ to make laboratory measurements of line profiles in N_2O and to detect thermal emission from Mars and from the Moon. This experiment was conducted at the coudé focus of the 30-inch telescope at the Goddard Optical Research Facility (Greenbelt, MD) in January and February of 1974.

In heterodyne detection, infrared radiation (possibly from a remote source) is mixed with the output of an intense coherent local oscillator and a signal is detected at the difference frequency (called the intermediate frequency or IF). The spectral characteristics of the remote source are preserved at the IF frequency, except that the frequency scale is effectively translated by an amount equal to the local oscillator frequency. Using the very best current infrared detectors and preamplifiers, the IF bandwidth can extend from nearly D.C. to beyond 1 GHz and radio detection techniques may be used to determine the fine structure of the spectrum of the remote source. The limiting spectral resolution is set by the spread in the local oscillator frequency which for semiconductor diode lasers can be less than 10^5 Hz .¹ Thus, spectral resolutions exceeding $1:10^8$ are possible, far in excess of the resolutions attainable with conventional infrared spectroscopic techniques.² Furthermore, the signal-to-noise ratio (S/N) of a heterodyne detection system can always approach the limit imposed by the photon statistics of the received signal (given sufficient local oscillator power) whereas other infrared detection techniques are limited by detector noise or background photon noise. Thus, heterodyne spectroscopy is ideally suited for the

identification of atomic and molecular species in remote infrared sources and for determination of the line profiles, giving information on the kinetic energy distributions, turbulence conditions, and doppler velocities of the sources. The extremely high spectral and spatial resolutions enable the study of low density, low kinetic temperature astronomical sources such as stars, comets, interstellar clouds, and the upper atmospheres of planets. A heterodyne system can also be operated with lesser resolution if the electronics following the photo-mixer integrate the intermediate frequency signal over the whole bandwidth. Even in that case, however, the spectral resolution of an instrument with a 1 GHz detector at $8.5 \mu\text{m}$ would be around 0.04 cm^{-1} .

Heterodyne radiometric methods using gas lasers as local oscillators have recently been successfully applied to detection of broad-band thermal radiation from astronomical sources³⁻⁷ and to laboratory detection of pollutant gases⁸. While these results demonstrated the feasibility of making such measurements, the real power of heterodyne detection lies in remote spectroscopic measurements at high resolution, long wavelength, and high sensitivity. Peterson et al.⁹ recently made the first heterodyne spectroscopic measurements of C^{13}O_2 absorption lines in the Mars atmosphere near $11 \mu\text{m}$. Their device featured a CO_2 local oscillator and so their observations were limited to the intermediate frequency bandwidth ($\sim 1 \text{ GHz}$) on either side of the discrete local oscillator frequencies. Obviously, the versatility of a heterodyne spectrometer is limited by the frequency range over which the laser local oscillator may be tuned and still have sufficient power and

coherence for heterodyne operation. Continuously tuneable (over several hundred wave numbers) lasers are not yet available in the middle infrared, but semi-tuneable lasers have recently become available.

Cryogenically cooled semi-tuneable semiconductor diode lasers^{1,10} have been proposed as possible local oscillators for remote heterodyne measurements of air pollutants by Hinkley and Kelley.¹¹ Diode lasers emitting at nominal wavelengths from 5-34 μm can now be manufactured¹² and are commercially available¹³. The nominal wavelength can be pre-selected to within 5 cm^{-1} of the desired value by varying the chemical composition of the ternary semiconductor compounds. These lasers generally show multimode output, with mode separations of up to several wave numbers. Each mode can be current tuned continuously over $\sim 1 \text{ cm}^{-1}$ ($\sim 3 \times 10^{10}$ Hz at 8.5 μm). Thus, tuning is truly continuous over each single mode and piece-wise continuous over the wavelength range of multi-mode operation. The multi-mode range is 10 cm^{-1} if only current tuning is used and 40 cm^{-1} if magnetic field tuning is available as well.

We have built a heterodyne spectrometer using PbSe semiconductor diode lasers as local oscillators, a HgCdTe photodiode as a photomixer,¹⁴ and an 8-channel filter bank as a line receiver. The spectrometer was interfaced with a computer-controlled 30-inch telescope, and a Dicke-type chopper was used to look alternately at the astronomical source and a precision black-body reference source (Fig. 1). An array of 4 current-tuned diode lasers provided adequate local oscillator power for direct absorption measurements over nearly the entire 1160 to 1190 cm^{-1} region,

however, the range over which the power was sufficient for heterodyne detection was considerably smaller ($\sim 5 \text{ cm}^{-1}$). The maximum total power output from our best diode was $\sim 2 \text{ mW}$ of which $\sim 100 \text{ }\mu\text{W}$ of single mode coherent power was actually incident on our photomixer. The mode structure of individual lasers and the current tuning characteristics were determined with an 0.5 m Ebert monochromator, using the photomixer as a direct detector.

For heterodyne detection the laser and source signals were superimposed by the beam-splitting lens and focussed onto the photomixer (Fig. 1). A 200 MHz band at the intermediate frequency output of the photodiode was then fed into the 8 channel filter bank and the output voltage from each 25MHz channel was linearly converted to a frequency which was counted with a multichannel analyzer for the period of integration. Data acquisition was synchronized with the chopping frequency. The measured IF noise-equivalent-power (NEP) for our system was $1.5 \times 10^{-19} \text{ W/Hz}$ at $10.6 \text{ }\mu\text{m}$ and $100 \text{ }\mu\text{W}$ local oscillator power.¹⁵ At $8.5 \text{ }\mu\text{m}$ and $100 \text{ }\mu\text{W}$ the extrapolated NEP is $3.6 \times 10^{-19} \text{ W/Hz}$. This is a factor of 2 higher than the theoretical limit ($\frac{h\nu}{\eta}$, η = quantum efficiency) and about a factor of 2 below the result derived from black body measurements at $8.5 \text{ }\mu\text{m}$. Neglecting optical losses and taking into account the marginal laser power, the minimum detectable power for our system for 8 minute integration times was $\sim 1 \times 10^{-16} \text{ W}$ in a 25 MHz bandwidth. The resolving power of our heterodyne spectrometer was 1.4×10^6 . A comparison of the resolution of our heterodyne spectrometer with other high resolution spectroscopic techniques used to measure remote sources near $10 \text{ }\mu\text{m}$ is given in Figure 2. A portion of the N_2O spectrum near $8.4 \text{ }\mu\text{m}$ is shown

along with the resolving bandwidth of the Michelson, tilting filter and heterodyne techniques. It is seen that only the heterodyne technique is capable of line profile measurements of the given 10 Torr N_2O lines.

The filter bank together with the tunability of our lasers enabled us to measure spectral line profiles in the heterodyne mode by positioning our local oscillator within 200 MHz of the line center.

Line profiles in the ν_1 band (100-000) of N_2O were measured directly and in the heterodyne mode. A cell filled with N_2O at 10 Torr pressure was placed in position 1 and the absorption line positions and profiles were determined by direct detection of tuned laser line absorption (Fig. 1). The cell was then placed in position 2 and the lines were heterodyne detected in absorption against a 1300°K black body continuum (the optical path to the telescope was blocked). The line was then moved through our filter bank by slightly tuning our local oscillator between measurements. The results of these measurements are shown in Figure 3. The broken line is the absorption line profile measured in the direct detection mode and the histogram corresponds to the line profile measured in the heterodyne mode in the 8 channel filter bank. The pressure broadened absorption linewidth was about 170 MHz. For comparison purposes, both lines were normalized to the same amplitude and the direct detected line was drawn at the predicted line center (PLC) position in the filter bank. The noise is greater in the heterodyne line profile because the power in a 25 MHz heterodyne channel corresponds to $\sim 10^{-13}$ W, whereas the direct measurements correspond to $\sim 10^{-4}$ W of laser power at $8.5 \mu m$. Good agreement is seen between the line shapes measured in the direct and

heterodyne modes and between the predicted and observed positions of the line center in the heterodyne mode (Figure 3). It is believed that this is the first time that a molecular absorption line profile has been measured using both active and passive measurement techniques.

The heterodyne system was next applied to astronomical observations and was used successfully to spectroscopically measure thermal radiation from Mars and from the Moon. We were unable to detect Jupiter, Venus, and Comet Kohoutek (1973f) in the heterodyne mode. Failure to observe Jupiter and Venus can be accounted for by their lower temperatures (125°K and 215°K respectively^{16,17}) and by their low positions in the sky during the observing runs. Both planets were always near the horizon where atmospheric absorption and air turbulence were extremely high. These factors would significantly decrease our signal-to-noise ratios. Comet Kohoutek was too faint in the visible during our period of observations for us to be able to align its image in our heterodyne system with sufficient accuracy.

The black body continuum near $8.5\text{ }\mu\text{m}$ from the Moon and from Mars gave heterodyne signal-to-noise ratios of 3 to 8. The variations were caused by daily changes in the local oscillator power and changing atmospheric conditions. Representative results are shown in Figure 4. The signal in each channel is represented by a bar whose height is proportional to the net flux received from the source. The huge signal in the last channel of the lunar data was probably a system noise pulse and was disregarded in determining the signal-to-noise ratio. The theoretical system signal-to-noise ratio for a black-body source is given by^{6,18}

$$S/N = \frac{\alpha \eta_{\text{eff}} (B\tau)^{1/2}}{(e^{h\nu/kT} - 1)}$$

where B is the IF bandwidth, τ the integration time and α the transmission of the atmosphere and optics. η_{eff} is the effective quantum efficiency of our photodiode and is equal to the true quantum efficiency degraded by a factor stemming from the lack of sufficient local oscillator power to reach the shot-noise limit. The true quantum efficiency of our photodiode at $8.5 \mu\text{m}$ was about 13%. η_{eff} was estimated to lie between 2 and 6.5% depending on laser operation.

Theoretical signal to noise ratios (i.e. with $\alpha = 1$ in eq. 1) based on local lunar temperature at the region of observations, known electronic parameters, and the appropriate η_{eff} were from 5 to 10 times greater than the experimentally obtained values, implying $\alpha = 0.1 \rightarrow 0.2$. System optical transmission at $8.5 \mu\text{m}$ was measured to be about 0.2 and atmospheric transmission¹⁷ at 50° zenith angle for this area can be as high as 80%; thus, the resulting α of 0.16 is consistent with the range of values obtained by comparing the experimental and theoretical S/N ratios discussed above.

The observed S/N ratios for Mars are more compatible with a temperature of 300°K than the expected $\sim 250^\circ\text{K}$.^{9,17} The effective quantum efficiency changed between the measurements of Moon and Mars due to greater laser output during the Mars run. The quantitative results are of only limited accuracy because of the frequent variation in laser power, poor telescope pointing stability, changes in zenith angle between runs, and daily changes in atmospheric conditions. Accurate monitoring

of all these parameters was not feasible at the time of the experiments.

The astronomical results obtained using diode lasers were compared with similar measurements made using a CO₂ laser. No attempt was made to optimize the system for 10.6 μm . The higher laser power enabled us to achieve $\eta_{\text{eff}} = 20\%$ at 10.6 μm . Signal-to-noise ratios for $\tau = 4$ minutes were from 1.5 to 9 depending on conditions. We found atmospheric transmission to be 0.24 for the Moon data and 0.47 for the Mars run using same day measurements on both sources, their expected temperatures, and estimated optical losses in our system. The factor of two is well within experimental uncertainty considering that the zenith angles were not identical.

Although we were able to obtain heterodyne signals with diode lasers as local oscillators, the measurements were far from straightforward. The coherent output power from presently available diode lasers is generally below that required for shot noise limited heterodyne operation. Only one of several diodes supplied had sufficient power output. Due to the large divergence of the laser output beam great difficulties were encountered in the design of the optical system to collect, focus, and match the laser output to our heterodyne field of view. The selection of the desired laser mode using a grating monochromator introduced intolerable losses in single-mode laser power and heterodyne measurements had to be made using all modes simultaneously. Many of these problems can be eliminated by using diode lasers of higher power output. Some success has already been achieved in this endeavor (e.g. Reference 19). The diode laser output was sensitive to nearly

all environmental effects, e.g. room temperature variations, mechanical and acoustic vibrations, liquid helium level, and heat sinking parameters of the helium dewar. Note, that the resolution of our instrument of 25 MHz corresponds at $8.5\text{ }\mu\text{m}$ to about 0.6 parts per million in relative wavelength. Environmental effects can easily cause physical changes of that order in diode dimensions, index of refraction, focussing optics, etc. Meaningful measurements could only be made when all the parameters remained stable during the period of data acquisition. Needless to say, conditions sometimes changed between measurements making accurate comparisons between runs difficult.

These results demonstrate the capability of our electronic and optical system and that heterodyning can be achieved using semiconductor diode laser local oscillators. We have shown the feasibility of spectroscopic observations of laboratory and astronomical sources with such an infrared heterodyne spectrometer. With proper design and improved diode lasers future devices should approach quantum noise limited operation.

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REFERENCES

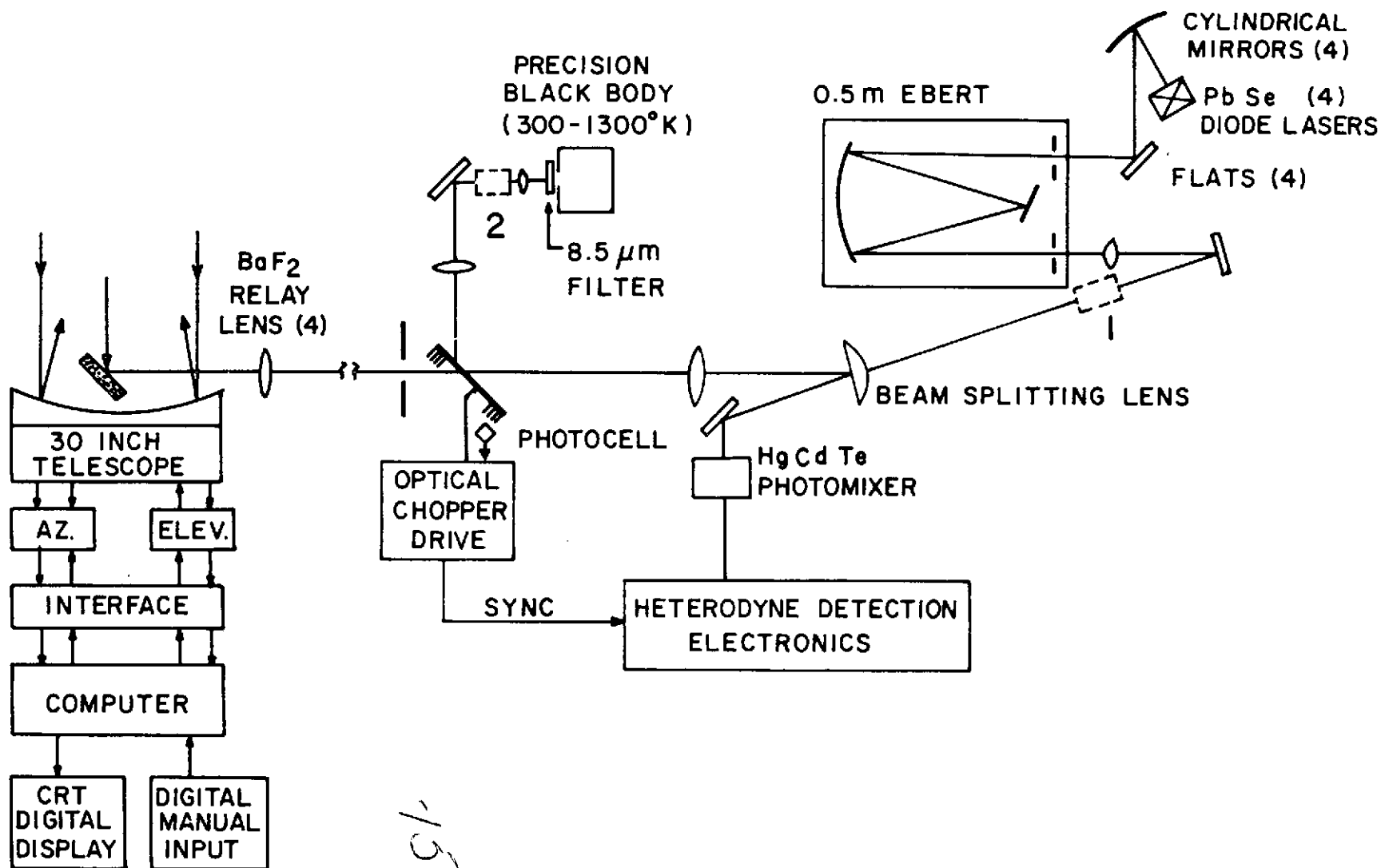
1. E. D. Hinkley, Appl. Phys. Letters, 16, 351 (1970).
2. $1:10^4$ is considered to be very high resolution near $10\text{ }\mu\text{m}$ using Michelson interferometers e.g. R. Hanel, M. Foreman, T. Meilleur, R. Wescott, J. Pritchard, Appl. Optics, 8, 2059 (1969). Tilting filter interferometers have been built which achieve resolutions approaching $1:10^5$, however, a separate set of filters is required for each line to be studied.
3. H. Nieuwenhuijzen, Mon. Not. R. Astro. Soc. 150, 325 (1970).
4. C. Veth, Th. de Graaw, J. C. Shelton, and H. van de Stadt, Astron. and Astrophys. 26, 479 (1973).
5. D. DeBatz, P. Granes, J. Gay, and A. Journet, Nature Phys. Sci. 245, 89 (1973).
6. Th. De Graaw and H. van de Stadt, Nature Phys. Sci. 246, 73 (1973).
7. A. J. DiNardo, B. J. Peyton, S. C. Cohen, J. H. McElroy, R. J. Coates, IEEE QE-9, 6, 662 (1973).
8. R. T. Menzies and M. S. Shumate, Science, 184, 570 (1974).
9. D. W. Peterson, M. A. Johnson, and A. L. Betz, private communication (submitted to Nature); see also M. A. Johnson, J. Opt. Soc. Am. 62, 1350 (1972).
10. G. A. Antcliff and S. G. Parker, J. Appl. Phys., 44, 4145 (1974).
11. E. D. Hinkley and P. L. Kelley, Science, 171, 635 (1971).
12. E. D. Hinkley, Opto-electronics, 4, 69 (1972).
13. A. D. Little, Inc., Cambridge Massachusetts.
14. The HgCdTe photodiode and matched low noise preamplifier were

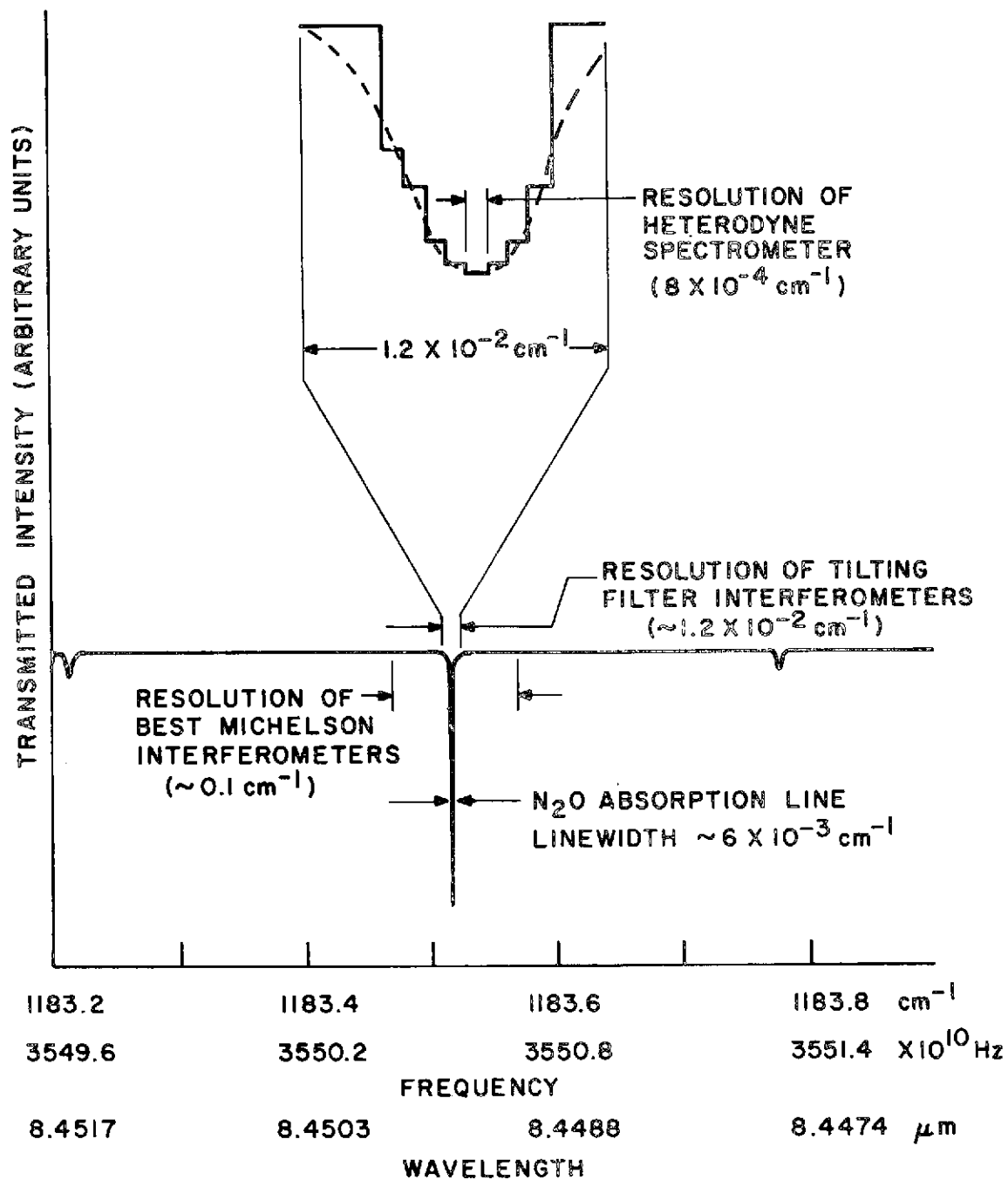
built and loaned to us by Mr. B. J. Peyton and his associates of Airborne Instrument Laboratories, Melville, N.Y. The requirements for designing, evaluating, and optimizing parameters in infrared heterodyne receivers are reviewed by B. J. Peyton, A. J. DiNardo, G. M. Kanischak, F. R. Arams, A. R. Lange, and F. W. Sard, IEEE QE-8, 2, 252 (1972).

15. Measurements were done at A.I.L.
16. F. C. Gillette, F. J. Low, and W. A. Stein, Ap. J., 157, 925 (1969).
17. V. Kunde, private communication.
18. J. H. McElroy, Appl. Optics, 11, 1619 (1972).
19. K. W. Nill, S. H. Groves, A. J. Strauss Post-Deadline paper at VIII International Quantum Electronics Conference, San Francisco, June 10 - 13, 1974.

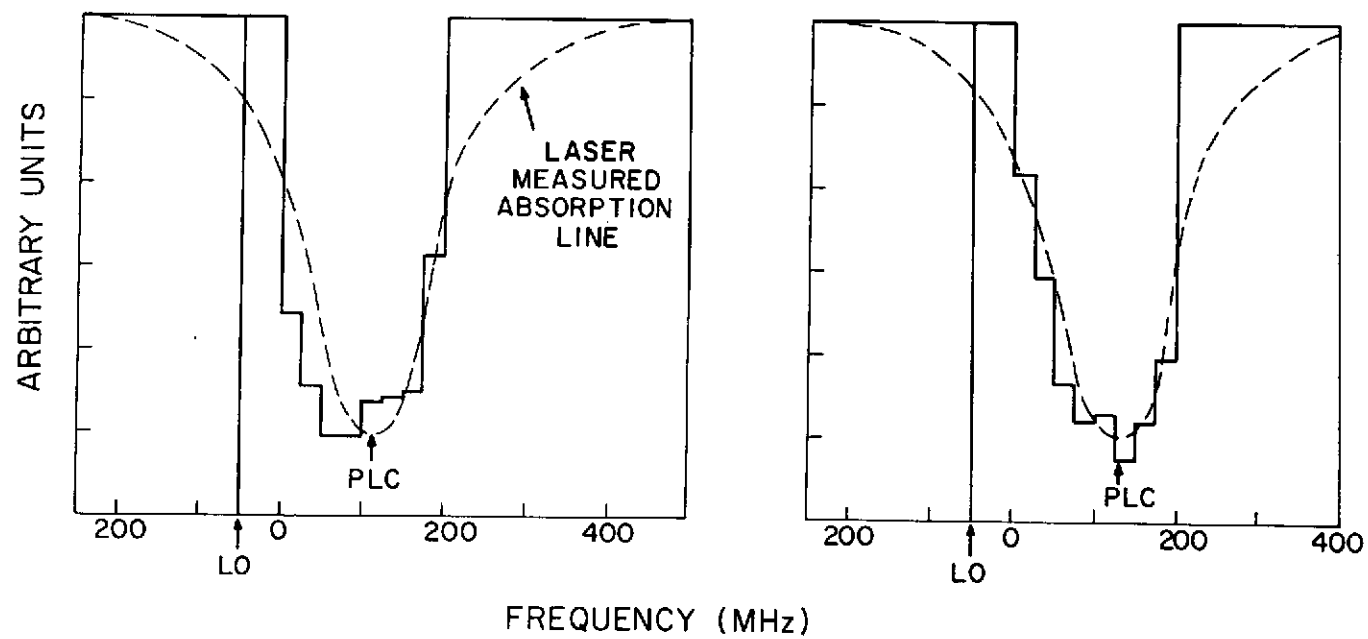
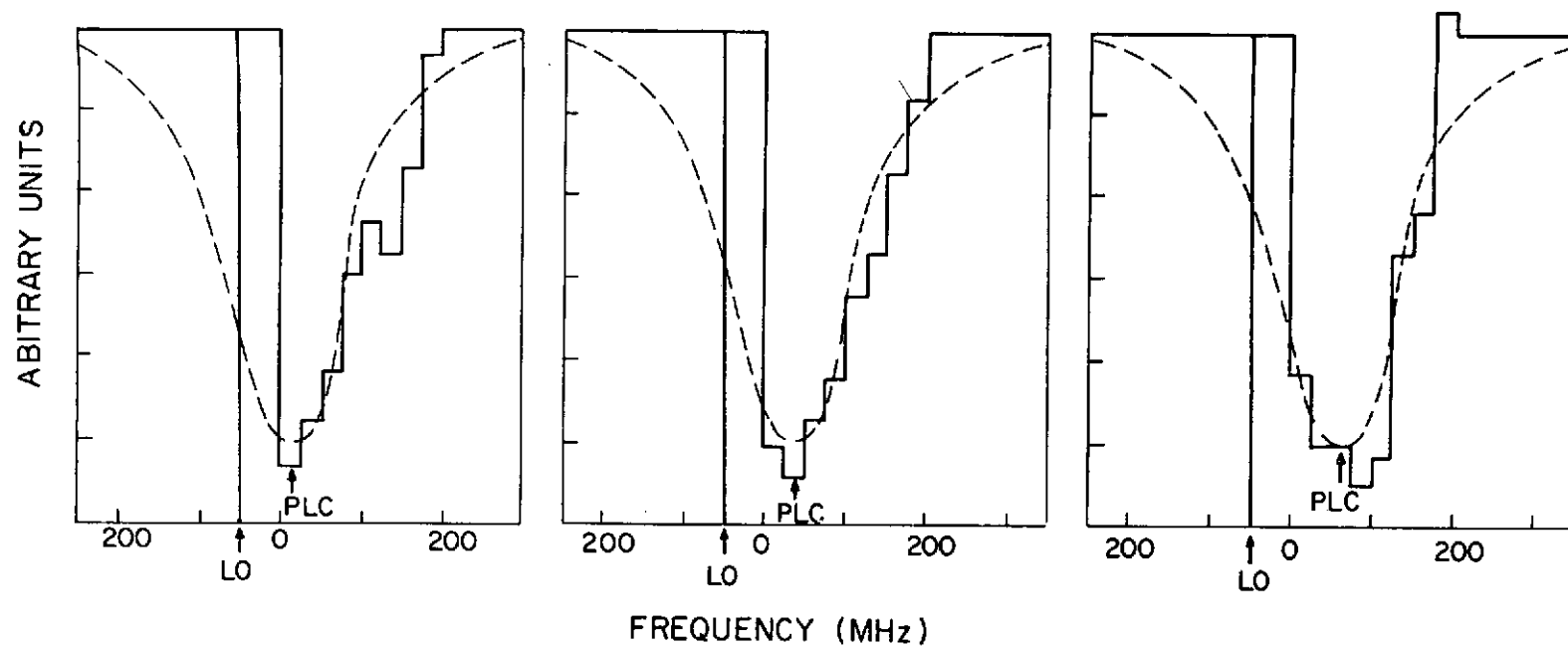
FIGURE CAPTIONS

- Figure 1. 8.5 μm Spectrometer Optical System.
- Figure 2. Comparison of Spectral Resolution of the Heterodyne with
Other High Resolution Techniques Near 8.5 μm .
- Figure 3. Heterodyne Detection of N_2O Absorption Line.
- Figure 4. Heterodyne Signals at 8.5 μm from the Moon and Mars.





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